



Atmospheric Environment 35 (2001) 3293-3302

ATMOSPHERIC ENVIRONMENT

www.elsevier.com/locate/atmosenv

Temporal trends of black carbon concentrations and regional climate forcing in the southeastern United States

J.-S. Im, V.K. Saxena*, B.N. Wenny

Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University, Raleigh, NC 27695-8208, USA

Received 7 July 2000; received in revised form 17 October 2000; accepted 30 October 2000

Abstract

The effect of black carbon (BC) on climate forcing is potentially important, but its estimates have large uncertainties due to a lack of sufficient observational data. The BC mass concentration in the southeastern US was measured at a regionally representative site, Mount Gibbes (35.78°N, 82.29°W, 2006 m MSL). The air mass origin was determined using 48-h back trajectories obtained from the hybrid single-particle Lagrangian integrated trajectory model. The highest average concentration is seen in polluted continental air masses and the lowest in marine air masses. During the winter, the overall average BC value was 74.1 ng m⁻³, whereas the overall summer mean BC value is higher by a factor of 3. The main reason for the seasonal difference may be enhanced thermal convection during summer, which increases transport of air pollutants from the planetary boundary layer of the surrounding urban area to this rural site. In the spring of 1998, abnormally high BC concentrations from the continental sector were measured. These concentrations were originating from a biomass burning plume in Mexico. This was confirmed by the observations of the Earth probe total ozone mapping spectrometer. The BC average concentrations of air masses transported from the polluted continental sector during summer are low on Sunday to Tuesday with a minimum value of 256 ng m⁻³ occurring on Monday, and high on Wednesday to Friday with a maximum value of 379 ng m⁻³ occurring on Friday. The net aerosol radiative forcing (scattering effects plus absorption effects) per unit vertical depth at 2006 m MSL is calculated to be -1.38×10^{-3} W m⁻³ for the southeastern US. The magnitude of direct radiative forcing by aerosol scattering is reduced by $15 \pm 7\%$ due to the BC absorption. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Aerosol; Black carbon; Absorption; Direct radiative climate forcing; Temporal patterns

1. Introduction

The carbonaceous component of atmospheric aerosols is composed of two main fractions (i.e., organic carbon and black carbon (BC)). Organic carbon is generally associated with condensed organic compounds which is primarily a scattering medium, whereas BC is generally defined as the absorbing component of carbonaceous aerosols. Absorption of solar radiation by BC heats the atmosphere while reducing solar irradiance at the

E-mail address: saxena@unity.ncsu.edu (V.K. Saxena).

ground, thereby altering the vertical temperature profile. The effects of BC on climate forcing (i.e., a warming influence on the atmosphere, by adding to greenhouse forcing or by decreasing cooling by the scattering components of aerosols), although recognized as potentially important, were estimated with large uncertainties (IPCC, 1994; Penner et al., 1994).

The BC is mainly produced by the incomplete combustion of fossil and biomass fuels. It is ubiquitous and can be found in soils, ice, sediments and the atmosphere. Wet and dry deposition are known as important sinks for BC (Hansen et al., 1988; Ogren et al., 1984). Since most of the BC particles are in the fine-particle size mode (e.g., Ogren and Charlson, 1983; Penner et al., 1992), dry removal rate is small (Ogren et al., 1984). Bahrmann and Saxena (1998) showed BC concentration decreased abruptly after

^{*}Corresponding author. Tel.: +1-919-515-7290; fax: +1-919-515-7802.

precipitation due to scavenging effects. Lifetime of BC is relatively short (40 h in rainy climates to 1 week in clean, dry regions) and controlled by several factors (e.g., the initial size distribution, the concentration of ambient particles, the frequency and duration of precipitation, and the efficiencies of removal mechanisms) (Ogren and Charlson, 1983). BC is found in the atmosphere in both urban and remote regions. In general, BC is assumed to be inert and non-volatile (Ogren and Charlson, 1983), thus the light absorbing aerosol component measured by optical method is commonly used as a good tracer for combustion (e.g., Hansen et al., 1988; Allen et al., 1999). Since the lifetime of the particles with diameters of greater than a few micrometers is quite short, it is necessary when using a global climate model or a regional climate model to concentrate on the fraction of the aerosols which is capable of long-range transport.

The presence of anthropogenic aerosol in a remote region may result in changes in the solar radiation balance. Therefore, the investigation of the seasonal variations and long-term trends in BC aerosol parameters as well as the indication of possible source region is important. In this study, we investigate the influence of longrange transport on measured BC mass concentrations at a representative mountain-top site in North Carolina, its seasonal and weekly patterns, and the effects of BC on the regional climate of the southeastern United States. The results could aid in understanding the meteorological transport of atmospheric aerosol and improve the knowledge of the climatology due to aerosol BC in the southeastern United States.

2. Methodology

2.1. Experimental setup

The experimental site (North Carolina State University Research Station) is located on the peak of Mt. Gibbes (35.78°N, 82.29°W, 2006 m MSL), in the Blue Ridge Mountains of western North Carolina. The research site lies approximately 4km southwest of Mount Mitchell (2038 m MSL), the highest peak in eastern North America. Detailed descriptions of the site are available in Bahrmann and Saxena (1998). The air mass origin was determined using 48-h back-trajectories obtained from the hybrid single-particle Lagrangian integrated trajectory (HY-SPLIT) model (Draxler and Hess, 1997). Air mass source regions were classified as polluted continental, continental, and marine with some continental influence, based on the SO_x , NO_x emission inventories obtained from the US Environmental Protection Agency (Fig. 1). (Detailed descriptions on the sector division can be found in Ulman and Saxena (1997).) In situations where the back trajectories crossed the boundaries between adjacent sectors, the classification of air

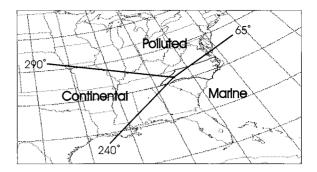


Fig. 1. Map of the United States illustrating the geographical location and classification of the different air mass types influencing the research sites.

mass history became somewhat subjective and was based on the perceived amount of time spent in any one sector. Since we assume the air mass arriving at the research site is transported from its source of origin, the classification of air mass does not imply a pure air mass but rather a modified air mass. Validation of the air mass sector classification was obtained through the analysis of cloud water pH (Ulman and Saxena, 1997) and the ionic concentration found in cloud water (Deininger and Saxena, 1997). Ulman and Saxena (1997) reported the lowest average pH value to originate in the polluted continental sector. Deininger and Saxena (1997) showed that cloudforming air masses from the polluted continental sector were abundant in sulfates. They also found that air masses from the marine sector were characterized by the presence of sea-salts, and air masses from the continental sector were characterized by the presence of calcium, although a major component in both marine and continental air masses was also sulfate.

2.2. Instrumentation

A Magee Scientific aethalometer was deployed at the site and provided realtime, continuous measurement of BC mass concentration. The measurements were recorded at 15 min intervals and were obtained for Spring (5/4–5/31, 1998), Summer (6/1–8/31, 1998), and Winter (11/11–12/31, 1997 and 12/17–12/31, 1998) to investigate the seasonal patterns of BC concentration.

The aethalometer is designed to measure the fraction of the carbonaceous aerosol that absorbs light over a broad region of the visible spectrum by determining the attenuation of the light transmitted through the sample when collected on a fibrous filter (Hansen, 1996). Gundel et al. (1984) have shown that optical absorption is proportional to BC mass concentration, such that BC mass concentration (g m⁻³) is equal to the measured absorption (i.e., absorption coefficient, m⁻¹) divided by the

absorption efficiency (m² g⁻¹). The value of absorption efficiency depends on the size of particles, the incident light wavelength, and the type of mixing between BC and scattering component (such as organic matter and sulfates) (e.g., Waggoner et al., 1981; Rosen and Hansen, 1984; Liousse et al., 1993; Chýlek et al., 1995; Martins et al., 1998). Several authors (e.g., Clarke et al., 1987; Japar et al., 1986) obtained values around $10 \,\mathrm{m}^2\,\mathrm{g}^{-1}$ for absorption efficiency of aerosol particles and Gundel et al. (1984) reported a value of 25 m² g⁻¹ by comparing direct thermal measurements of ambient BC with measurements of light attenuation. In addition, Martins et al. (1998) reported absorption efficiencies measured for various types of biomass fires ranging between 5.2 and $19.3 \,\mathrm{m^2 \, g^{-1}}$ with an average value of $12.1 \pm 4.0 \,\mathrm{m^2 \, g^{-1}}$. The value used in our aethalometer is 19 m² g⁻¹ and can be considered as first-step approximation in determining BC mass concentration in our study. Internally mixed particles composed of an absorbing core surrounded by a non-absorbing shell have, in general, a greater BC mass absorption efficiency than pure BC particles or externally mixed particles (e.g., Ackerman and Toon, 1981; Chýlek et al., 1995). This is because the non-absorbing shell increases the total cross-sectional area of the particles and focuses light towards the absorbing core, causing the same amount of BC in the internally mixed structure to absorb more than pure BC particles (Martins et al., 1998).

The aerosol scattering coefficient was also measured from 15 July to 13 August 1998. A Radiance Research M-903 nephelometer measures the total scattering coefficients at a wavelength of 530 nm, using the geometry of a standard integrating nephelometer under ambient relative humidity (RH). With general meteorological values (i.e., pressure, temperature, and RH), the values of total scattering coefficient are monitored and recorded as 15 min averages. Calibration is accomplished by adjusting

the span so that the indicated scattering coefficient matches the calculated value when sampling calibration span gas at local pressure and temperature conditions. Particle-free air and freon gas (CHClF₂) are used as zero reference and span gas, respectively. The optical and electrical background noise (usually wall scatter) is sufficiently low to allow the measurement of total scattering coefficient from less than $10^{-6} \, \mathrm{m}^{-1}$ to greater than $10^{-3} \, \mathrm{m}^{-1}$.

3. Results and discussions

3.1. Seasonal patterns of BC concentrations

To investigate the influence of long-range transport on measured BC mass concentrations, each BC sample was categorized by air-mass-type. The monthly average for each air-mass-type is shown in Fig. 2. Except for May 1998, the highest average BC concentration values were found in polluted continental air masses and the lowest BC values were found in marine air masses. BC mass concentrations in summer 1998 agree well to those in summer 1996, 1997 (Bahrmann and Saxena, 1998). The seasonal variation of BC mass concentrations was also investigated (Fig. 3). During the summer season, the mean BC mass concentration is 327.4 ng m⁻³ for polluted continental air masses, 165.9 ng m⁻³ for continental air masses, and 103.3 ng m⁻³ for marine air masses. During the winter season, the mean BC mass concentration is 107.4 ng m⁻³ for polluted continental air masses, 86.7 ng m⁻³ for continental air masses, and 28.1 ng m⁻³ for marine air masses. In general, BC mass concentrations observed in other areas in winter are higher than those in summer, due to domestic and commercial heating in urban areas (e.g., $1.74 \,\mu g \, m^{-3}$ for winter compared to 0.54 µg m⁻³ for summer in Nova Scotia, Canada

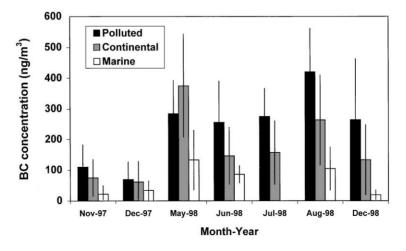


Fig. 2. Average monthly BC concentrations for each sector at Mount Gibbes, NC.

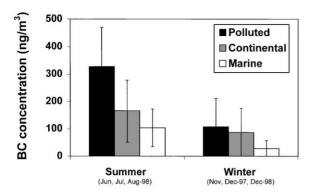


Fig. 3. Average BC concentrations for each sector during the summer months (June–August in 1998) and during the winter months (November and December in 1997 and December in 1998).

(Chýlek et al., 1999) and $2.0 \, \mu g \, m^{-3}$ for winter compared to 1.6 µg m⁻³ for summer over the Washington DC area (Malm et al., 1994). However, rural regions such as our research site located at a higher altitude, are influenced by long-range transport of air pollutants, and show different seasonal patterns. One of the main reasons for the seasonal difference is the enhanced thermal convection during the summer. This may increase transport of planetary boundary layer air pollutants from the surrounding urban area to this rural area. Therefore, the mixing height in the site can be as high as or higher than 2006 m MSL during the summer months. On the other hand, we would expect lower mixing height in the winter. Hence, during the winter, the air on Mt. Gibbes is cleaner, with an overall average value of 74.1 ng m⁻³, whereas the overall summer mean value is higher by about a factor of 3. These results are in good agreement with the seasonal variations of daily median BC mass concentrations at a high-alpine research station during 1995-1997 (Lavanchy et al., 1999).

In the spring of 1998, abnormally high BC concentrations from the continental sector were measured (Fig. 2). The monthly mean value of BC (375.0 ng m⁻³) in May 1998 for continental air masses is even greater than that of polluted continental air masses (283.9 ng m⁻³) and about twice as high as the concentrations in the continental air masses of May, 1997. A maximum daily average BC concentration is also shown in this continental air mass (May 16, 1998) with a value of 673 ng m^{-3} . This anomaly can be explained by a massive biomass burning in Central America and southern Mexico in the spring of 1998. The aerosols originating from the forest-fire smoke plume in Mexico were transported to the southeastern US. The plume was also detected by the Earth-Probe total ozone-mapping spectrometer (TOMS) (NASA/GSFC, Laboratory for Atmospheres; http://toms.gsfc.nasa.gov/) and the most extensive smoke coverage at the research site occurred on May 16 (Fig. 4).

Therefore, these BC measurements obtained from continental sector air masses during May 1998 is evidence of the long-range transport of BC produced by biomass burning in Mexico.

3.2. Weekly patterns of BC concentration

To investigate day-of-week distribution of BC concentrations for the air masses transported from the polluted continental sector, 48-h back-trajectories arriving at the research site at 1300 EST during June and July 1998 are drawn in Fig. 5. The BC concentration is averaged for three Sundays, two Mondays, one Tuesday, two Wednesdays, two Thursdays, two Fridays, and two Saturdays displayed in Fig. 5 to illustrate the day-of-week distribution (Fig. 6). The mean BC concentration increases from Monday to Friday and decreases from Friday to Monday. The day-of-week distribution for the polluted continental air masses is also calculated for the entire summer period in order to generalize BC weekly patterns (Fig. 7). This shows the mean BC concentration of the week along with 95% confidence intervals for the estimated mean values. As can be seen, the BC concentrations are low on Sunday to Tuesday with a minimum value of $256\pm10\,\mathrm{ng}\,\mathrm{m}^{-3}$ occurring on Monday and higher from Wednesday to Friday with a maximum value of $379 \pm 13 \,\mathrm{ng}\,\mathrm{m}^{-3}$ occurring on Friday. Considering the transport time required from northern urban areas, the minimum occurring on Monday can represent lower BC emissions on Saturday and Sunday in the polluted source regions. The higher BC concentrations shown on Wednesday to Friday may be due to weekday pollution emissions in the source regions. The summer average of the BC concentration temporal (day-of-week) distribution measured at our site and that measured in southwestern Pennsylvania (Allen et al., 1999) are compared in Table 1. The major air pollution sources in the latter region were reported as the Hatsfield electric generating station, vehicular emissions, and open burning of residential and commercial trash allowed on Saturdays and/or Wednesdays (Allen et al., 1999). It is apparent that there is about a 1-day lag for peak and minimum BC concentrations at our site compared to one of BC emission source regions in the polluted sector. The overall average BC concentration at our site (322 ng m⁻³) is reduced to about 25% of that in southwestern PA (1270 ng m⁻³). Although this comparison is insufficient to explain the exact mean time for transport to our site from southeastern PA, it can be used to approximate the time lag of the weekly BC concentration trend and BC amount transported from a source region.

3.3. Comparison with scattering measurements

Total aerosol scattering coefficient measurements at the site were compared with the BC absorption coefficient

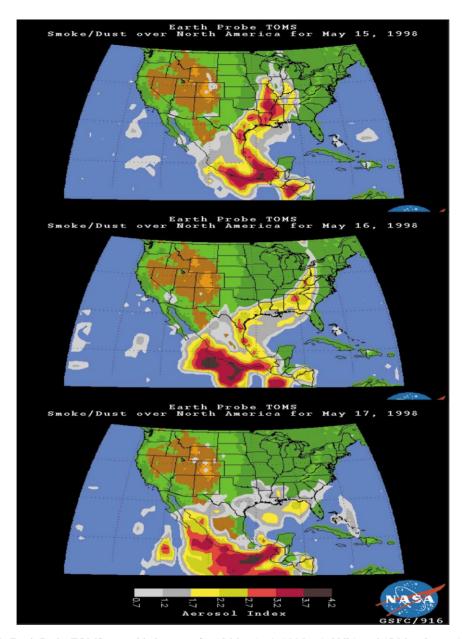
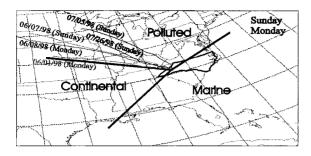
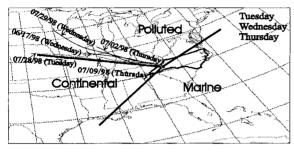


Fig. 4. Earth-Probe TOMS aerosol index maps for 15 May (top), 16 May (middle), and 17 May (bottom) 1998.

measurements. Scattering by aerosols is strongly dependent on the RH at which it is measured, due to the hygroscopic growth nature of most atmospheric aerosols (e.g., Tang and Munkelwitz, 1977; Malm et al., 1994; Hegg et al., 1996; Kotchenruther et al., 1999). At our research site, the maximum value of total scattering is observed just before sunrise and the minimum value is observed near 2 PM, which is coincident with the trend of RH. In order to eliminate the diurnal variation of total scattering, daily average values were

calculated. Fig. 8 illustrates the daily means of total scattering and absorption coefficients from 15 July to 13 August 1998. Although the magnitude of the absorption coefficients is less than that of total scattering coefficients at the site, the daily mean absorption coefficients correlate well (correlation coefficient, R=0.86) with the daily mean total scattering coefficients. This suggests that the transported aerosols responsible for scattering and absorption have originated from the same emission sources.





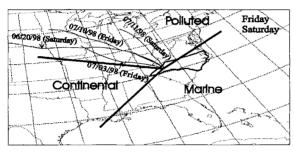


Fig. 5. Forty-eight-hour back-trajectories for polluted continental air mass cases arriving at the research site at 1300 EST during June and July 1998.

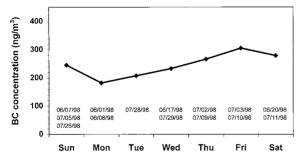


Fig. 6. Temporal (day-of-week) distribution of black carbon concentrations for the trajectories displayed in Fig. 5. Each point represents the average concentration for 1000 EST to 1600 EST during the days indicated.

3.4. BC effects on direct radiative climate forcing

Numerous studies of the direct effect of tropospheric sulfate aerosols have shown an aerosol impact on climate

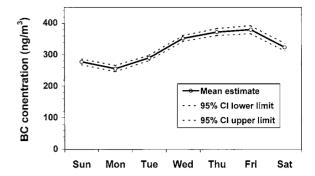


Fig. 7. Temporal (day-of-week) distribution of black carbon concentrations averaged for polluted continental air masses during the summer period (June–August, 1998). The line with circles denotes the mean value and dashed lines denote 95% confidence interval (CI) for the mean estimate.

through reflection of solar radiation back to space (e.g., Charlson et al., 1991, 1992; Kiehl and Briegleb, 1993; Boucher and Anderson, 1995; Saxena and Menon, 1999). Estimates of the globally averaged annual direct radiative forcing due to anthropogenic sulfates range from $-0.3 \, {\rm W \, m^{-2}}$ (Kiehl and Briegleb, 1993) to $-1.3 \, {\rm W \, m^{-2}}$ (Charlson et al., 1992). The direct cooling effects of aerosols are reduced by the presence of BC (e.g., Chýlek and Wong, 1995; Chýlek et al., 1995). The BC within the sulfate aerosol reduces the expected sulfate direct cooling effect by about 0.034 W m⁻² for each 1% of the BC to sulfate mass mixing ratio (Chýlek et al., 1995). Haywood and Shine (1995) reported that fossil-fuel-derived BC caused positive global mean radiative forcing ranged from +0.03 to +0.24 W m⁻² for an assumed BC/sulfate mass ratio of between 0.05 and 0.1.

With direct measurements of optical properties for aerosols in the southeastern US, aerosol direct radiative forcing is calculated using the equation obtained by Chýlek and Wong (1995):

$$\Delta F_{\rm R} = \, - \, \frac{S_{\rm o}}{4} T_{\rm atm}^2 (1 - A_{\rm c}) [(1 - R_{\rm s})^2 2B \tau_{\rm sc} - 4R_{\rm s} \tau_{\rm abs}], (1)$$

where $S_{\rm o}$ is the solar constant, $T_{\rm atm}$ is the atmospheric transmission, $A_{\rm c}$ is the fractional cloud amount, $R_{\rm s}$ is the surface albedo of the underlying surface, B is the fraction of average daily solar radiation scattered back to space (i.e., upscatter fraction computed from backscatter fraction), and $\tau_{\rm sc}$ and $\tau_{\rm abs}$ are the optical depth of the aerosol layer due to light scattering and absorption, respectively.

To calculate τ_{sc} and $\tau_{abs},$ the following equations were used:

$$\tau_{\rm sc} = \int_{Z_1}^{Z_2} \sigma_{\rm sp} \mathrm{d}z,\tag{2}$$

$$\tau_{abs} = \int_{Z_1}^{Z_2} \sigma_{ap} dz, \tag{3}$$

Table 1
Comparison of temporal (day-of-week) distribution of black carbon concentrations at Mount Gibbes, NC, to those obtained in southwestern PA. The numbers represent mean + standard deviation

Day of week	Polluted air masses arriving at Mt. Gibbes NC summer of 1998 BC concentration (ng m ⁻³)	Air masses in southwestern PA summer of 1990 (Allen et al., 1999) BC concentration (ng m ⁻³)
Sunday	278 ± 107	647 ± 338
Monday	256 ± 108	1016 ± 271
Tuesday	290 ± 109	1294 ± 453
Wednesday	352 ± 130	1490 ± 600
Thursday	372 ± 157	1612 ± 486
Friday	379 ± 161	1409 ± 561
Saturday	324 ± 149	1421 ± 969

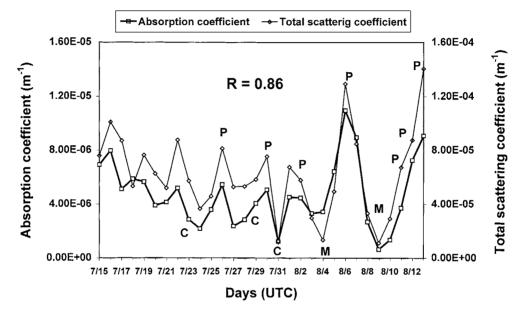


Fig. 8. Daily means of absorption coefficient and scattering coefficient measured from 15 July to 13 August 1998. P, C, and M denote polluted continental, continental, and marine air mass influences for specific times, respectively, based on 48 h back trajectory analyses that were available during the period.

where $\sigma_{\rm sp}$ and $\sigma_{\rm ap}$ are the total scattering coefficient and absorption coefficient, respectively. To compare contributions of aerosol scattering and absorption to regional climate at a certain height, the values of $\tau_{\rm sc}$ and $\tau_{\rm abs}$ were calculated for unit vertical depth at 2006 m MSL. Parameters used to calculate the direct radiative forcing in this study were $S_{\rm o}=1370~{\rm W~m^{-2}}$, $T_{\rm atm}=0.76$, $A_{\rm c}=0.61$, and $R_{\rm s}=0.15$ based on the values used in the box model (Charlson et al., 1992). In addition, the value of B used in the computation of $\Delta F_{\rm R}$ was estimated using the regression equation of backscatter fraction ($\beta=-0.0005\times{\rm RH}+0.125$) obtained from three-wavelength nephelometer at a nearby research station (35.66°N, 82.38°W, 951 m MSL) and Henyey–Greenstein phase function (Wiscombe and Grams, 1976) to be 0.23 at

RH = 68%. This RH is the mean ambient RH during the measurement and is consistent with the clear sky daytime RH (68.6%) in the eastern United States during July, obtained by Boucher and Anderson (1995). Direct radiative forcing per unit vertical depth (W m $^{-3}$) was computed for aerosol scattering and absorption at 2006 m MSL of the southeastern US during 15 July–13 August 1998. The net aerosol direct radiative forcing (scattering effects plus absorption effects) per unit vertical depth was calculated to have a negative value of -1.38×10^{-3} W m $^{-3}$. The magnitude of direct radiative forcing by aerosol scattering is reduced by 15 \pm 7% due to the BC absorption. Thus, the presence of BC does not significantly change the estimates of aerosol direct cooling effect in the southeastern US. A regional cooling trend in

the mean maximum surface temperature over the southeastern US from 1949 to 1994 (Saxena and Yu, 1998) is potentially an observational confirmation for the estimate.

Haywood and Shine (1995) expressed the aerosol forcing as a function of optical depth:

$$\Delta F = -DS_{\rm o} T_{\rm atm}^2 (1 - A_{\rm c}) \omega_{\rm o} B \delta [(1 - R_{\rm s})^2 - \frac{2R_{\rm s}}{B} \left(\frac{1}{\omega_{\rm o}} - 1\right)], \tag{4}$$

where δ is the optical depth, D is the fractional daylength (the value of D=0.5 is used in this study), and $\omega_{\rm o}$ is single scatter albedo (the ratio of scattering to absorption plus scattering for a thin aerosol layer). The parameters $(S_{\rm o}, T_{\rm atm}, A_{\rm c}, B, {\rm and} R_{\rm s})$ are the same as those defined in Eq. (1). Rearranging the Eq. (4), the aerosol radiative forcing per unit optical depth (aerosol radiative forcing efficiency) is obtained as follows:

$$\frac{\Delta F}{\delta} = -DS_o T_{\text{atm}}^2 (1 - A_c) \omega_o B [(1 - R_s)^2 - \frac{2R_s}{B} \left(\frac{1}{\omega_o} - 1\right)].$$
(5)

The direct radiative forcing per unit optical depth are calculated as a function of single scatter albedo during 15 July-13 August 1998. In this computation, the value of 0.23 for upscatter fraction is used as mentioned previously. At our experimental site, 93% of the calculated single scatter albedo are in the range of 0.90-0.96 and the overall average direct radiative forcing per unit optical depth is calculated to be $-20 \pm 2 \,\mathrm{W \, m^{-2}}$. The magnitude of this cooling estimate is smaller than that for dry aerosols in the range of -23 to -33 W m⁻² over central and eastern North America (Sheridan and Ogren, 1999). It is partially due to the aerosol particle size growth with RH in our case considering the hygroscopic growth factor. That is, the smaller upscatter fraction value used in our study is responsible for this difference. Neglecting BC absorption effects (i.e., single scatter albedo = 1), the direct radiative forcing per unit optical depth is derived to be $-25.6 \,\mathrm{W}\,\mathrm{m}^{-2}$ which approaches to the value of $-32 \,\mathrm{W}\,\mathrm{m}^{-2}$ obtained by Charlson et al. (1991) when the value of 0.29 for upscatter fraction is used. In our study considering the effects of BC absorption and aerosol hygroscopic growth nature, the BC reduces the expected sulfate direct cooling effect per unit optical depth by $21 \pm 9\%$.

4. Summary and conclusion

The BC mass concentrations in the southeastern United States were measured at a regionally representative site near Mount Mitchell, North Carolina, the highest peak in eastern North America. The results obtained are the following:

- During the experiment, except for May 1998, the highest average BC concentration is seen in polluted continental air masses and the lowest in marine air masses
- (2) During the winter the air is cleaner with an overall average value of 74.1 ng m⁻³, whereas the overall summer mean value is higher by about a factor of 3. The main reason for the seasonal difference may be enhanced thermal convection during summer, which increases transport of planetary boundary layer air pollutants from the surrounding urban area to this rural site.
- (3) In the spring of 1998, abnormally high BC concentrations from the continental sector were measured. These concentrations were originating from biomass burning from the forest-fire smoke plume in Mexico. This was confirmed by the observations of the Earth-Probe TOMS.
- (4) The mean BC concentrations of air masses transported from the polluted continental sector during summer are low from Sunday to Tuesday with a minimum value of 256 ng m⁻³ on Monday and higher from Wednesday to Friday with a maximum value of 379 ng m⁻³ on Friday.
- (5) The daily mean BC absorption and scattering coefficients are well correlated (R = 0.86), which suggests the aerosols originated from the same emission sources.
- (6) The net aerosol radiative forcing (scattering effects + absorption effects) per unit vertical depth at 2006 m MSL was calculated to be $-1.38\times 10^{-3}\,\mathrm{W\,m^{-3}}$ for the southeastern United States. The magnitude of direct radiative forcing by aerosol scattering is reduced by $15\pm7\%$ due to the BC absorption.

The effect of BC on climate forcing is important because of its high absorption of solar radiation, but the effect is estimated with a large uncertainty due to a lack of sufficient observational data. Therefore, the investigation of the seasonal variations and long-term trends in BC aerosol parameters as well as the indication of possible source area is important. The results obtained from continuous in situ measurements of BC aid in understanding the meteorological transport of atmospheric aerosol and improve the knowledge of the climatology due to aerosol BC in the southeastern United States.

Acknowledgements

This research was supported by NASA's Mission to Planet Earth (MTPE) under Contract #NAS1-18944

from Langley Research Center, Hampton, VA and US EPA's STAR Grant #R-825248. The authors acknowledge Bill Barnard for his helpful comments concerning this research.

References

- Ackerman, T.P., Toon, O.B., 1981. Absorption of visible radiation in atmosphere containing mixtures of absorbing and nonabsorbing particles. Applied Optics 20, 3661–3668.
- Allen, G.A., Lawrence, J., Koutrakis, P., 1999. Field validation of a semi-continuous method for aerosol black carbon (aethalometer) and temporal patterns of summertime hourly black carbon measurements in southwestern PA. Atmospheric Environment 33, 817–823.
- Bahrmann, C.P., Saxena, V.K., 1998. The influence of air mass history on black carbon concentration in the southeastern United States. Journal of Geophysical Research 103, 23153–23161.
- Boucher, O., Anderson, T.L., 1995. General circulation model assessment of the sensitivity of direct radiative forcing by anthropogenic sulfate aerosols to aerosol size and chemistry. Journal of Geophysical Research 100, 26117–26134.
- Charlson, R.J., Langner, J., Rodhe, H., Leovy, C.B., Warren, S.G., 1991. Perturbation of the Northern Hemisphere radiative balance by back scattering from anthropogenic sulfate aerosols. Tellus 43B, 152–163.
- Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley Jr., J.A., Hansen, J.E., Hofmann, D.J., 1992. Climate forcing by anthropogenic aerosols. Science 255, 423–430.
- Chýlek, P., Wong, J., 1995. Effect of absorbing aerosols on global radiation budget. Geophysical Research Letters 22, 929–931.
- Chýlek, P., Videen, G., Nat, D., Pinnick, R.G., Klett, J.D., 1995. Effect of black carbon on the optical properties and climate forcing of sulfate aerosols. Journal of Geophysical Research 100, 16325–16332.
- Chýlek, P., Kou, L., Johnson, B., Boudala, F., Lesins, G., 1999. Black carbon concentrations in precipitation and near surface air in and near Halifax, Nova Scotia. Atmospheric Environment 33, 2269–2277.
- Clarke, A.D., Noone, K.J., Heinzenberg, J., Warren, S.G., Covert, D.S., 1987. Aerosol light absorption measurement techniques: analysis and intercomparisons. Atmospheric Environment 21, 1455–1465.
- Deininger, C.K., Saxena, V.K., 1997. A validation of back trajectories of air masses by principal component analysis of ion concentrations in cloud water. Atmospheric Environment 31, 295–300.
- Draxler, R.R., Hess, G.D., 1997. Description of the HYSPLIT_4 modeling system. NOAA Tech Memo ERL ARL-224, December, 24pp. Web address: http://www.arl.noaa.gov/ready/ hysplit4.html, NOAA Air Resources Laboratory, Silver Spring, MD.
- Gundel, L.A., Dod, R.L., Rosen, H., Novakov, T., 1984. The relationship between optical attenuation and black carbon concentration for ambient and source particles. Science of the Total Environment 36, 197–202.

- Hansen, A.D.A., 1996. Magee Scientific Aethalometer User's Guide. Magee Sci. Company, Berkeley, CA, 56 pp.
- Hansen, A.D.A., Bodhaine, B.A., Dutton, E.G., Schnell, R.C., 1988. Aerosol black carbon measurements at the South Pole: initial results 1986–1987. Geophysical Research Letters 15, 1193–1196
- Haywood, J.M., Shine, R.P., 1995. The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation budget. Geophysical Research Letters 22, 603-606.
- Hegg, D.A., Covert, D.S., Rood, M.J., Hobbs, P.V., 1996. Measurement of aerosol optical properties in marine air. Journal of Geophysical Research 101, 12893–12903.
- IPCC, Climate Change, 1994. In: Houghton J.T. et al. (Eds.), Radiative Forcing of Climate Change and an Evaluation of the IPCC 1S92 Emission Scenarios. Cambridge University Press, Cambridge, New York.
- Japar, S.M., Brachaczek, W.W., Gorse, R.A., Norbeck, J.M., Pierson, W.R., 1986. The contribution of elemental carbon into the optical properties of rural atmospheric aerosols. Atmospheric Environment 20, 1281–1289.
- Kiehl, J.T., Briegleb, B.P., 1993. The radiative roles of sulfate aerosols and greenhouse gases in climate forcing. Science 260, 311-314.
- Kotchenruther, R.A., Hobbs, P.V., Hegg, D.A., 1999. Humidification factors for atmospheric aerosols off the mid-Atlantic coast of the United States. Journal of Geophysical Research 104, 2239–2251.
- Lavanchy, V.M.H., Gäggeler, H.W., Nyeki, S., Baltensperger, U., 1999. Elemental carbon and black carbon measurements with a thermal method and an aethalometer at the highalpine research station Jungfraujoch. Atmospheric Environment 33, 2759–2769.
- Liousse, C., Cachier, H., Jennings, S.G., 1993. Optical and thermal measurement of black carbon aerosol content in different environments: variation of the specific attenuation cross-section sigma (σ). Atmospheric Environment 27, 1203–1211.
- Malm, W.C., Sisler, J.F., Huffman, D., Eldred, R.A., Cahill, T.A., 1994. Spatial and seasonal trends in particle concentration and optical extinction in the United States. Journal of Geophysical Research 99, 1347–1370.
- Martins, J.V., Artaxo, P., Liousse, C., Reid, J.S., Hobbs, P.V., Kaufman, Y.J., 1998. Effects of black carbon content, particle size, and mixing on light absorption by aerosols from biomass burning in Brazil. Journal of Geophysical Research 103, 32041–32050.
- Ogren, J.A., Charlson, R.J., 1983. Elemental carbon in the atmosphere: cycle and lifetime. Tellus 35B, 241–254.
- Ogren, J.A., Groblicki, P.J., Charlson, R.J., 1984. Measurement of the removal rate of elemental carbon from the atmosphere. Science of the Total Environment 36, 329–338.
- Penner, J.E., Dickinson, R.E., O'Neill, C.A., 1992. Effects of aerosol from biomass burning on the global radiation budget. Science 256, 1432–1434.
- Penner, J.E., Charlson, R.J., Hales, J.M., Laulainen, N.S., Leifer, R., Novakov, T., Ogren, J., Radke, L.F., Schwartz, S.E., Travis, L., 1994. Quantifying and minimizing uncertainty of climate forcing by anthropogenic aerosols. Bulletin of the American Meteorological Society 75, 375-400.

- Rosen, H., Hansen, A.D., 1984. Role of combustion generated carbon particles in the absorption of solar radiation in Arctic haze. Geophysical Research Letters 11, 461–464.
- Saxena, V.K., Yu, S.-C., 1998. Searching for a regional fingerprint of aerosol radiative forcing in the southeastern US. Geophysical Research Letters 25, 2833–2836.
- Saxena, V.K., Menon, S., 1999. Sulfate-induced cooling in the southeastern US: an observational assessment. Geophysical Research Letters 26, 2489–2492.
- Sheridan, P., Ogren, J.A., 1999. Observations of the vertical and regional variability of aerosol optical properties over central and eastern North America. Journal of Geophysical Research 104, 16793–16805.
- Tang, I.N., Munkelwitz, H.R., 1977. Aerosol growth studies III: ammonium bisulfate aerosols in a moist atmosphere. Journal of Aerosol Science 8, 321–330.
- Ulman, J.C., Saxena, V.K., 1997. Impact of air mass histories on the chemical climate of Mt. Mitchell, North Carolina. Journal of Geophysical Research 102, 25451–25465.
- Waggoner, A.P., Weiss, R.E., Ahlquist, N.C., Covert, D.S., Will, S., Charlson, R.J., 1981. Optical characteristics of atmospheric aerosols. Atmospheric Environment 15, 1891–1909.
- Wiscombe, W.J., Grams, G.W., 1976. The backscattered fraction in two-stream approximations. Journal of Atmospheric Science 33, 2440–2451.